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DETONATION PRESSURE MEASUREMENTS ON PETN

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Abstract. PETN is widely recognized as an example of nearly ideal detonation performance. The chemical composition is such that little or no carbon is produced in the detonation products. The reaction zone width is less than currently detectable. (<1 ns) Observations on PETN have thus become a baseline for EOS model predictions. It has therefore become important to characterize the detonation parameters as accurately as possible in order to provide the most exacting comparisons of EOS predictions with experimental results.

We undertook a painstaking review of the detonation pressure measurements reported in an earlier work that was presented at the Fifth Detonation Symposium and found that corrections were required in determining the shock velocity in the PMMA witness material. We also refined the impedance calculation to account for the difference between the usual "acoustic" method and the more accurate Riemann integral. Our review indicates that the CJ pressures previously reported for full density PETN require an average lowering of about 6 percent. The lower densities require progressively smaller corrections.

We present analysis of the records, supporting hydrodynamic simulations, the Riemann integral results, and EOS parameter values derived from the revised results.

Introduction

This work is a reanalysis of work in "Equation of State of Detonation Products" by Hornig, et al[1] as presented at the Fifth Detonation Symposium. There were three major corrections to the original presentation. The first had to do with reducing the data using the quadratic equation as given in the paper, which was originally applied with the quadratic term on the distance instead of the time variable. This resulted in artificially elevated values for the shock velocity in the PMMA and also concealed a systematic offset in the 0,0 point.

The second correction was to ignore entirely the 0,0 point in the data reduction. For reasons to be explained, this point is thought to have a systematic error. This approach resulted in an average calculated CJ pressure that was about 5 percent lower than that reported for the higher density experiments.

The third correction was to determine the CJ pressure from the measured shock velocity in the PMMA reference material by means of the Riemann integral instead of the acoustic approximation. This lowered the CJ pressure slightly more than one percent for the highest PETN density experiments.

The quadratic extrapolation to determine entering shock velocity in the PMMA and the Riemann correction were verified by simulating the experiment with a very high resolution computer calculation where the input CJ pressure was set at 31.5GPa,the PETN density at 1.765grams per cubic centimeter, and the detonation velocity at 8.274 km/sec. The equation of state used for the PMMA was the same as used in the data reduction. The three corrections applied to the computer-generated results yielded a value of P_{CJ} of 31.4 GPa, well within probable error.

In order to obtain continuous values for $dD/d\rho_0$ we modified the formula for fitting the detonation velocity as a function of density. The modified formula shown here retain the previous values to within 0.5%.

$D=3.30-3.7(\rho_0-.4) - 3(\rho_0 - 1.6)^2$ for $\rho_0 > 1.6$

$D=3.30-3.7(\rho_0-.4)$ for $.4< \rho_0 <1.6$

$D=3.30-3.7(\rho_0-.4) + 3.5(\rho_0 - .4)^2$ for $\rho_0 < .4$

These fitted results were sed in combination with the revised Chapman Jouguet ressure results to yield the Chapman Jouguet derivative parameters.

The Fifth Symposium paper included observations at charge densities .27 to .48 g/cc using optical techniques. There were large deviations in the data. In addition the results for pressures and derivative values in most cases exceeded realistic limits. Judging that reliable results at these lower densities must await future experiments we have not included these results in this paper.

Experimental description

The work is described in detail in the referenced paper[1]. The PETN detonation pressure was obtained from the shock velocity transmitted to a reference material (PMMA) by a plane detonation wave in the PETN. In that paper, the CJ pressure was then determined by use of the acoustic approximation. The measurements reviewed here were those where the shock velocity in the PMMA was obtained by using the shock electric effect described by Hayes[2] to observe the shock arrival time at each interface of a stack of five accurately measured discs (1.27 mm thick).

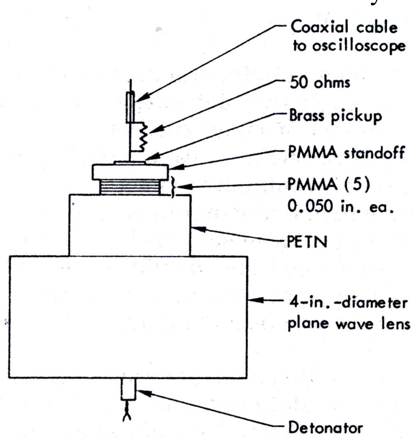

Figure 1. Experimental arrangement for P_{CJ} measurement by shock electric effect.

Evaluation of the PMMA shock measurements

When the x,t data points from the experiments on high density PETN were properly fit to quadratic equations, it was discovered that there was a systematic offset in the 0,0 points. That is, there is a constant in the fit on the order of 7 nanoseconds. This means that at x=0 mm the time has already progressed 7 nanoseconds. This is a substantial error and had to be resolved. The method chosen was to simulate the experiment with a very high-resolution computer calculation. Then, the x,t data generated was used to back calculate the input pressure to the computer with the quadratic equation being used to calculate the entering shock velocity in the PMMA. The computer-generated data was fit perfectly by the quadratic equation with no significant value to the constant. When the 0,0 point was taken out of the calculation, the entering shock velocity in the PMMA was the same as with it in. From this, it was concluded that there was an error in the values of the experimental 0,0 points and all of the experimental data was reduced omitting the 0,0 points. An examination of the actual shock electric output from the experiment gave a possible clue to the problem. The signals from the shock transit of the PMMA interfaces were all very large and of one polarity while the signal from the shock entering the PMMA from the explosive was very small and of the opposite polarity. It is our belief that the offset seen in the 0,0 point arises from this divergence in signal strength and polarity to the oscilloscope used to record the data in the experiment. There is also a different anomaly that the reanalysis brought to light. For the highest density PETN samples, the ones that are only 12.7 mm thick yield a CJ pressure statistically lower than the 25.4 mm thick samples. One can either accept all of the data or come up with a reason why some should be rejected. It is our opinion that the results from the 12.7 mm samples should be rejected. This is because it is quite likely that the CJ pressures measured for the 12.7mm samples are low because of the very steep Taylor wave that follows the extremely thin reaction zone of the detonating PETN. If all values were to be averaged, the CJ pressure would be 31.5 GPa.

while the PETN CJ pressure from 25.4 mm thick samples averages 32.0 GPa if an errant data point not included in Table 1 is omitted.

Acoustic vs Riemann P_{CJ} calculation

The customary method for obtaining the value of the Chapman Jouguet pressure from shock wave impedance measurements is to apply the "acoustic" approximation to the shock wave observations as the means to connect the shock response of the witness material to the shock wave CJ state in the detonation products. Even for relatively low impedance witness materials such as the PMMA in these experiments it is generally assumed that the rarefaction of the detonation products from the CJ state follows a linear path in the P,u_P plane which is the negative of the slope of the hugoniot locus of the products. This is in fact the first term in the expansion of the Riemann result.

$P = P_{CJ} + (dP/du)(u- u_{CJ}) + .5(d^2P/du^2)(u - u_{CJ})^2 \ldots\ldots$

This approximation to the exact Riemann integral result is quite adequate for witness materials whose shock impedance and particle velocity closely matches that for the detonation products. For the highest density PETN charges the results from the Riemann integral analysis shows a measurable correction to the acoustic result. The Riemann integral is derived from the relation

$du/d\rho = - (C/\rho)_S$ where C is the sound speed (1)

The integral, in general, must be evaluated by iterative technique since the equation for C

$$C^2 = (dP/d\rho)_S \qquad (2)$$

$$dP/du =- C\rho \qquad (3)$$

leads to an expression which cannot be evaluated in closed form for all but simplest expressions for the isentropic pressure P_S.

The derivative (dP/du) is obtained from the JWL form of the equation of state for PETN at $\rho o = 1.762$ to calculate C. The integral was carried out over the interval V(CJ detonation products) to V(witness PMMA state) by transforming (1) from $C(\rho)$ to C(V) .

$u_P(PMMA) - u_P(CJ) = Int [(1/Vo)(-dP/dV)]^{1/2}dV/V \quad V = v/vo \qquad (4)$

The acoustic estimate is given by

$$Pcj = (P_{PMMA} + \rho_o Du_{PMMA})/2 \qquad (5)$$

The deviation of the Riemann result from the acoustic result is illustrated in figures 2 and 3. For PETN ($\rho_0 = 1.762$) the correction is - 4.0 kb. The correction to be applied at the lower densities can be determined by noting the deviation of the Riemann integral values for the pressure from the acoustic values and accounting for the slopes at the intercept as illustrated in figures 2 and 3. The deviation in pressure from the acoustic result can be calculated to within 1% as $\lambda(u_PPMMA - ,u_PPETNCJ)^2/2$ where $\lambda = 2.04$. This is found to apply for the higher densities. For the lower density PETN charges due to decrease in both the particle velocity difference and the coefficient .lthe deviation becomes negligible.

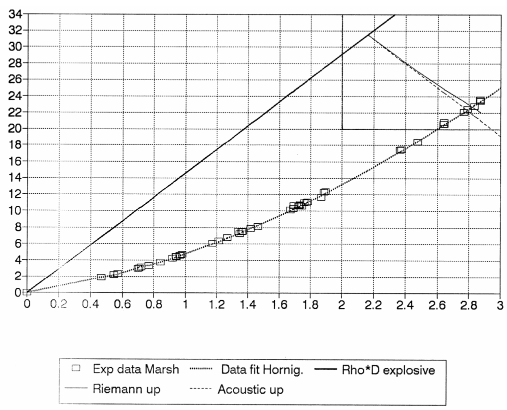

Figure 2. Comparison of Riemann and acoustic approximation.

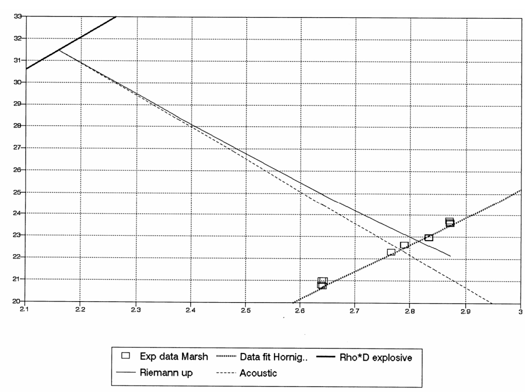

Fig. 3 Comparison of Riemann and acoustic approximation.

Adiabatic Γ parameter

The Γ values for the adiabatic derivative $\Gamma = (\mathrm{dlnP}/\mathrm{dlnV})_S$ are calculated as

$$\Gamma_{CJ} = P_{CJ}/(\rho_o D^{2)} - 1$$

Results of the calculations are plotted in Fig. 4.

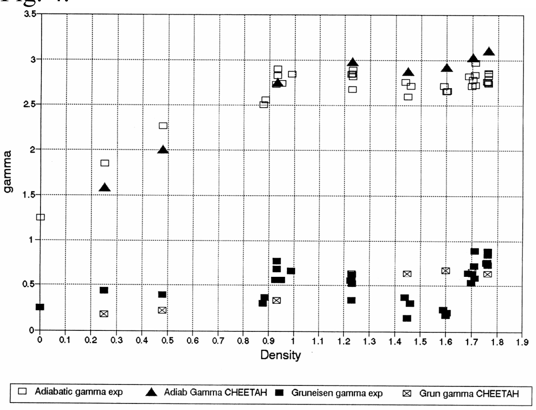

Fig. 4 PETN CJ derivatives.

Gruneisen γ parameter

The well known Jones relation provides a means to calculate the Gruneisen parameter for the CJ states at various densities.

γ is defined as $V(dP/dE)_V$

$\gamma = \Gamma(\Gamma-1-2Z)/\Gamma-Z)$

Γ is the adiabatic derivative at the CJ state

Z is the logarithmic derivative of the detonation velocity = $\mathrm{dlnD}/\mathrm{dln}\rho_0$

We are thus able to derive the dependence, $\gamma\rho_0)$ and since,

$\rho_0/\rho_{CJ} = \Gamma/(\Gamma+1)$

we can likewise derive the dependence $\gamma(\rho_{CJ})$ shown in Figure 4.

Pressure vs. density

Figure 5 summarizes and Table 1 lists the results of our recalculations.

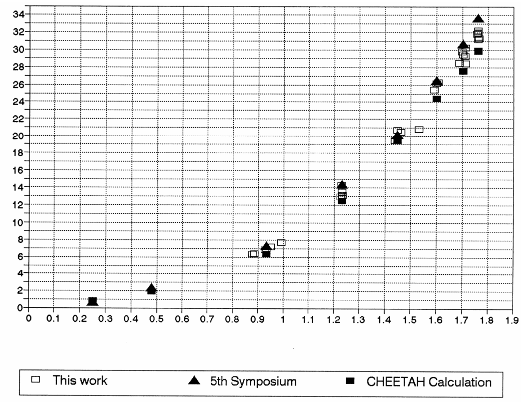

Fig.5 Comparison of 5th Det. Symp. values of PETN CJ pressure vs. density with recalculated values and values from Cheetah calculations.

Summary

Our revised results for Chapman Jouguet pressures are in almost all cases lower than previous results. The values for adiabatic gamma are thus, in general, greater than earlier estimates. Some comparisons with molecular model code calculations show that for this “baseline” explosive the molecular model yields values for pressures lower by approximately 7% than our results as illustrated in figure 5. Thus, although, as compared to the earlier results the current revised results for pressure are closer agreement with model predictions, the differences are still significant. The values obtained for the thermodynamic derivatives, Γ and γ are roughly in agreement with the trend of results from the CHEETAH calculations. The agreement with detonation velocities is very nearly within experimental variation (no shown here). Thus the deviations in comparisons for Γ and for γ vary mainly from the pressure deviations.

We believe that the current results can, with confidence, be used as a test comparison with calculational models.

Acknowledgments

The authors are indebted to Craig Tarver and David Hare for their patience and perseverance in setting up and running computer simulations of the detonation electric effect experiments on high density PETN.

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Table 1. Comparison of recalculated values of PETN P_{CJ} with values from 5th Detonation Symposium.

PETN Density (g/cc)	Detonation Velocity (km/sec)	DIMENSIONS Diam. x Length (mm)	5th DET. SYMPOSIUM[1] PMMA u_S (km/sec)		RECALCULATED VALUES PMMA u_S (km/sec)		
			PMMA u_S (km/sec)	PETN P_{CJ} (GPa)	PMMA u_S (km/sec)	P_{CJ} acoustic (GPa)	P_{CJ} Rm int. (GPa)
1.764	8.266	50.8 x 12.7	6.980	33.4	6.812	30.26	29.88
1.763	8.263	50.8 x 12.7	7.010	33.8	6.825	31.71	31.32
1.763	8.263	25.4 x 12.7	6.980	33.3	6.673	31.56	31.16
1.763	8.263	25.4 x 12.7	7.010	33.8	6.799	31.83	31.43
1.763	8.263	50.8 x 25.4	7.030	34.0	6.898	32.16	31.76
1.762	8.261	50.8 x 25.4	6.990	33.5	6.877	32.41	32.01
1.762	8.261	50.8 x 25.4	7.070	34.3	6.883	32.59	32.18
1.758	8.250	25.4 x 25.4	6.980	33.3	6.866	32.35	31.95
1.712	8.108	25.4 x 25.4	6.810	30.7	6.610	28.77	28.45
1.711	8.106	25.4 x 25.4	6.860	31.1	6.795	30.10	29.76
1.710	8.103	25.4 x 25.4	8.830	30.9	6.704	28.73	28.39
1.703	8.082	25.4 x 25.4	6.840	30.8	6.732	30.56	30.21
1.697	8.063	25.4 x 25.4	6.830	30.6	6.775	29.63	29.29
1.686	8.030	25.4 x 25.4	6.830	30.4	6.661	29.78	29.44
1.604	7.755	25.4 x 25.4	6.570	26.6	6.574	26.56	26.32
1.600	7.740	25.4 x 25.4	6.600	26.6	6.569	26.45	26.21
1.591	7.706	25.4 x 25.4	6.520	25.9	6.489	25.65	25.41
1.532	7.488	25.4 x 25.4	6.210	22.5	6.013	21.05	20.86
1.460	7.222	25.4 x 25.4	5.990	19.8	6.102	20.63	20.49
1.448	7.178	25.4 x 25.4	6.160	20.8	6.160	20.86	20.74
1.439	7.144	25.4 x 25.4	6.050	19.9	6.006	19.65	19.53
1.232	6.378	25.4 x 25.4	5.450	13.6	5.366	13.16	13.13
1.231	6.375	25.4 x 25.4	5.420	13.4	5.316	12.86	12.83
1.230	6.371	25.4 x 25.4	5.470	13.7	5.343	13.01	12.97
1.230	6.371	25.4 x 25.4	5.560	14.2	5.448	13.61	13.58
1.228	6.364	25.4 x 25.4	5.540	14.1	5.578	14.34	14.32
1.227	6.360	25.4 x 25.4	5.560	14.2	5.338	12.95	12.92
0.949	5.331	25.4 x 12.7	4.930	8.5	4.623	7.21	7.21
0.990	5.483	25.4 x 12.7	4.910	8.7	4.687	7.75	7.75
0.931	5.265	25.4 x 25.4	4.770	7.7	4.535	6.74	6.74
0.877	5.065	25.4 x 25.4	4.630	6.8	4.534	6.43	6.42
0.886	5.098	25.4 x 25.4	4.700	7.1	4.535	6.49	6.47
0.927	5.250	25.4 x 38.1	4.610	7.0	4.567	6.85	6.84
0.932	5.268	25.4 x 38.1	4.680	7.3	4.506	6.63	6.62

References

1. H. C. Hornig, E. L. Lee, M. Finger, and J. E. Kurrle, “Equation of State of Detonation Products”, *Fifth Symposium (International) on Detonation*, Pasadena, CA, Aug. 18-21 1970, pp.503-511 (1970).

2. B. Hayes, “The Detonation Electric Effect,” J.Appl. Phys., Vol. 38, pp. 507-511, Feb. 1967.

3. B. Hayes, ‘Electric Signals Generated by Shock and Detonation Waves,” presented at the Western States section of the Combustion Institute, Oct. 28, 1968.

4. L. E. Fried, K. R. Glaesemann, W. M. Howard, P. C. Souers, and P. A. Vtello, Cheetah Code, version 4.0, UCRL-CODE-155944, 2005.

Plus additional references from the original paper.